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Applicant(s)

Kwang-Leong CHOY and Bo SU

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### PRELIMINARY AMENDMENT

Assistant Commissioner for Patents Box Patent Application Washington, D.C. 20231

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Sir:

Prior to examination, please amend the application without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents, as follows:

### IN THE SPECIFICATION:

Please amend the specification without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents, as follows:

Page 1, line 1, insert --This application is a Continuation-in-Part of PCT/GB00/00013, filed January 5, 2000, designating the U.S., published July 29, 2000 as WO 00/42234 and claiming priority from GB 9900955.7 filed January 15, 1999. All of the above-mentioned applications, as well as all documents cited herein and documents referenced or cited in documents cited herein, are hereby incorporated herein by reference.

Page 9, please cancel paragraphs 9 and 10.

Page 11, please cancel the last paragraph.

Page 12, please cancel the first paragraph.

Page 12, paragraph 2, please amend as follows: -- Figure 7 illustrates an X-ray diffraction pattern of a BaZrO<sub>3</sub>; film as fabricated by Example 1;--

Page 12, paragraph 3, please amend as follows: -- Figures 8(a) and (b) illustrate surface and cross-sectional SEM micrographs of a CdS film as fabricated by Example 2; and --

Page 12, paragraph 4, please amend as follows: -- Figures 9(a) and (b) illustrate surface and cross-sectional SEM micrographs of a porous SiO<sub>2</sub> film as fabricated by Example 3.--

Page 18, please cancel lines 5 through 32.

Please cancel pages 19 through 21.

Page 22, please cancel lines 1 through 13.

Page 22 to 23, last paragraph, please amend as follows: -- A non-aqueous precursor solution for the deposition of a BaZrO<sub>3</sub> film was first prepared as follows. Barium metal (as supplied by Aldrich) was completely dissolved in a volume of 2-methoxyethanol (as supplied by Aldrich) by stirring at room temperature to form a barium alkoxide solution. A stoichiometric

amount of zirconium n-propoxide, a 70 wt% solution in n-propanol (as supplied by Aldrich), was then added to the barium methoxyoxide solution and refluxed at 124 °C, the boiling point of 2-methoxyethanol, for five hours. Then, a volume of 2-methoxyethanol was added to the refluxed solution to provide a 0.05 M precursor solution. Using the apparatus of the first-described embodiment and the so-prepared solution, a BaZrO<sub>3</sub> film was deposited on a silver substrate 5, with a substrate temperature of 600 °C, a substrate 5 to nozzle unit 11 distance of 30 mm, an electric field voltage of 10 kV, the piezoelectric transducer 43 of the aerosol generator 25 being operated at a frequency of 1.7 MHz and power of 50 W, and nitrogen being supplied at 30 ml per minute as the carrier gas. Nitrogen was used as the carrier gas to minimise the reaction between the barium and carbon dioxide in the air. The resulting film. formed in a single run without the need for any post-deposition heat treatment, was a crystalline BaZrO<sub>3</sub> film as characterized by the X-ray diffraction pattern illustrated in Figure 7.--.

Page 23, the first full paragraph, please amend as follow: -- A 0.01 M aqueous precursor solution for the deposition of a CdS film was first prepared using cadmium chloride and thiourea. Using the apparatus of the second-described embodiment and the so-prepared solution, a CdS film was deposited on a glass substrate 105, with a substrate temperature of 450 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 50 W, a deposition time of five minutes, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a dense, crystalline CdS film having a thickness of about 1 um, with a columnar structure and a smooth and uniform surface. SEM micrographs of the resulting film are illustrated in Figures 8(a) and (b). --.

Page 23, the second full paragraph, please amend as follows: -- A colloidal silica solution (Ludox<sup>TM</sup>, as supplied by DuPont) was diluted with distilled water to prepare an aqueous precursor solution having a concentration of 0.1 g/ml for the deposition of a SiO<sub>2</sub> film. Using the apparatus of the second-described embodiment and the so-prepared solution, a SiO<sub>2</sub> film was deposited on a glass substrate 105, with a substrate temperature of 200 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 20 W, a deposition time of one minute, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a porous SiO<sub>2</sub> film with a reticular structure. SEM micrographs of the resulting film are illustrated in Figures 11(a) and (b). --.

Page 24, first paragraph, please amend as follows: -- Finally, it will be understood that the present invention has been described in its preferred embodiments and can be modified in many different ways within the scope of the invention as defined by the appended claims. For example, in coating substrates 5, 105 of large area or complex geometric shape, the nozzle units 11, 111 could be modified to include a plurality of outlet ports 18, 118 or the film deposition apparatus could be modified to include a plurality of nozzle units 11, 111.--.

Page 24, after the first paragraph, please insert:

- -- The invention will now be further described by the following numbered paragraphs.
- 1. A method of depositing material, preferably a film, on a substrate, comprising the steps of:

providing a substrate;

heating the substrate;

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is attracted towards the substrate.

- 2. The method of paragraph 1, wherein the substrate is heated to a temperature of less than about 1050 °C, preferably less than about 800 °C.
- 3. The method of paragraph 1 or 2, wherein the substrate is heated during deposition,
- 4. The method of paragraph 3, wherein the thermal environment is such as to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 5. The method of any of paragraphs 1 to 4, wherein the material solution is an aqueous solution.
- 6. The method of any of paragraphs 1 to 4, wherein the material solution is a non-aqueous solution.
- 7. The method of any of paragraphs 1 to 6, wherein the aerosol droplets are at least partially charged prior to exiting the at least one outlet.
- 8. The method of paragraph 7, wherein the aerosol droplets are charged prior to

- exiting the at least one outlet.
- 9. The method of any of paragraphs 1 to 7, wherein the aerosol droplets are at least partially charged after exiting the at least one outlet.
- 10. The method of any of paragraphs 1 to 9, wherein the aerosol droplets are charged by the at least one electrode.
- 11. The method of any of paragraphs 1 to 10, wherein the at least one electrode is disposed at least partially in each aerosol flow.
- 12. The method of any of paragraphs 1 to 11, wherein the at least one electrode extends upstream of the at least one outlet.
- 13. The method of any of paragraphs 1 to 12, wherein the at least one electrode comprises an elongate element.
- 14. The method of any of paragraphs 1 to 13, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 15. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a single tip.
- 16. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a plurality of tips.
- 17. The method of any of paragraphs 1 to 16, wherein the nozzle unit includes a tubular section upstream of each outlet.
- 18. The method of paragraph 17. wherein the tubular section is an elongate section.
- 19. The method of paragraph 17 or 18, wherein the tubular section is a linear section.
- 20. The method of any of paragraphs 17 to 19, wherein the tubular section is substantially cylindrical.

- 21. The method of any of paragraphs 17 to 20, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 22. The method of any of paragraphs 17 to 21, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 23. The method of any of paragraphs 1 to 22, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 24. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by entraining the aerosol in a flow of a carrier gas fed to the nozzle unit.
- 25. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by applying a reduced pressure to the at least one outlet so as to entrain the aerosol in a flow of a carrier gas drawn through the nozzle unit.
- 26. The method of paragraph 24 or 25, wherein the carrier gas is a gas reactive to the material solution.
- 27. The method of paragraph 24 or 25, wherein the carrier gas is a gas non-reactive to the material solution.
- 28. The method of any of paragraphs 24 to 27 when appendant upon paragraph 4, wherein the flow of the carrier gas is provided such as to maintain the decreasing, temperature gradient.
- 29. The method of any of paragraphs 1 to 28, wherein the aerosol is delivered to the substrate such as to achieve a film growth rate of at least 0.2 um per minute, preferably at least 1 um per minute, more preferably at least 2 um per minute.
- 30. The method of any of paragraphs 1 to 29, wherein the flow rate through the at least one outlet is at least 5 ml per minute, preferably at least 50 ml per minute.

- 31. The method of any of paragraphs 1 to 30, wherein the nozzle unit is configured such that the directed aerosol flow from the at least one outlet is directed upwards, preferably substantially vertically upwards.
- 32. The method of any of paragraphs 1 to 31, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 33. The method of any of paragraphs 1 to 32, wherein the applied voltage is less than about 35 kV, preferably less than about 20 kV.
- 34. The method of any of paragraphs 1 to 33, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 35. The method of any of paragraphs 1 to 34, wherein the substrate is held stationary relative to the nozzle unit.
- 36. The method of any of paragraphs 1 to 34, further comprising the step of moving the nozzle unit relative to the substrate.
- 37. The method of paragraph 36, wherein the substrate is rotated, tilted and/or translated relative to the nozzle unit.
- 38. The method of any of paragraphs 1 to 37, when performed at atmospheric pressure.
- 39. The method of any of paragraphs 1 to 37, when performed below atmospheric pressure.
- 40. The method of any of paragraphs 1 to 37, when performed above atmospheric pressure.

- 41. An apparatus for depositing material, preferably a film, on a substrate, comprising:
  - a substrate holder for holding a substrate;
  - a heater for heating the substrate;
  - an aerosol generator for generating an aerosol comprising droplets of a material solution;
  - a charge applicator for applying a positive or negative charge to the aerosol droplets;
  - a nozzle unit in communication with the aerosol generator for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and a high voltage supply for generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is in use attracted towards the substrate.
- 42. The apparatus of paragraph 41, where configured to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 43. The apparatus of paragraph 41 or 42, wherein the at least one electrode extends upstream of the at least one outlet.
- 44. The apparatus of any of paragraphs 41 to 43, wherein the at least one electrode comprises an elongate element.
- 45. The apparatus of any of paragraphs 41 to 44, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.

- 46. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a single tip.
- 47. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a plurality of tips.
- 48. The apparatus of any of paragraphs 41 to 47, wherein the nozzle unit includes a tubular section upstream of each outlet.
- 49. The apparatus of paragraph 48, wherein the tubular section is an elongate section.
- 50. The apparatus of paragraph 48 or 49, wherein the tubular section is a linear section.
- 51. The apparatus of any of paragraphs 48 to 50, wherein the tubular section is substantially cylindrical.
- 52. The apparatus of any of paragraphs 48 to 51, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 53. The apparatus of any of paragraphs 48 to 52, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 54. The apparatus of any of paragraphs 48 to 53, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 55. The apparatus of any of paragraphs 41 to 54, further comprising a gas supply unit in communication with the aerosol generator for supplying a flow of a carrier gas for entraining the aerosol and delivering the same through the nozzle unit.
- 56. The apparatus of any of paragraphs 41 to 55, wherein the at least one outlet is directed upwards, preferably substantially vertically upwards.

- 57. The apparatus of any of paragraphs 41 to 56, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 58. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are held in fixed relation.
- 59. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are configured so as to be movable relative to one another.
- 60. The apparatus of paragraph 59, wherein the substrate is rotatable, tiltable and/or translatable relative to the nozzle unit.
- 61. The apparatus of any of paragraphs 41 to 60, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 62. A method of fabricating a powder, preferably an ultrafine powder, comprising the steps of:

providing a heated zone:

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to-form a powder.

63. An apparatus for fabricating a powder, preferably an ultrafine powder, comprising:

a heater for providing a heated zone;

an aerosol generator for generating an aerosol comprising droplets of a material solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and

a high voltage supply for generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is in use attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to form a powder.--

## IN THE CLAIMS:

Please amend the claims without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents, as follows:

1. A method of depositing material, preferably a film, on a substrate, comprising the steps of:

providing a substrate;

heating the substrate;

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is attracted towards the substrate.

- 2. The method of claim 1, wherein the substrate is heated to a temperature of less than about 1050 °C, preferably less than about 800 °C.
- 3. (Amended) The method of claim 1, wherein the substrate is heated during deposition,
- 4. The method of claim 3, wherein the thermal environment is such as to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 5. (Amended) The method of claim 1, wherein the material solution is an aqueous solution.
- 6. (Amended) The method of claim 1, wherein the material solution is a non-aqueous solution.

- 7. (Amended) The method of claim 1, wherein the aerosol droplets are at least partially charged prior to exiting the at least one outlet.
- 8. The method of claim 7, wherein the aerosol droplets are charged prior to exiting the at least one outlet.
- 9. (Amended) The method of claim 1, wherein the aerosol droplets are at least partially charged after exiting the at least one outlet.
- 10. (Amended) The method of claim 1, wherein the aerosol droplets are charged by the at least one electrode.
- 11. (Amended) The method of claim 1, wherein the at least one electrode is disposed at least partially in each aerosol flow.
- 12. (Amended) The method of claim 1, wherein the at least one electrode extends upstream of the at least one outlet.
- 13. (Amended) The method of claim 1, wherein the at least one electrode comprises an elongate element.
- 14. (Amended) The method of claim 1, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 15. (Amended) The method of claim 1, wherein the distal end of the at least one electrode includes a single tip.
- 16. (Amended) The method of claim 1, wherein the distal end of the at least one electrode includes a plurality of tips.
- 17. (Amended) The method of claim 1, wherein the nozzle unit includes a tubular section upstream of each outlet.

- 18. (Amended) The method of claim 17, wherein the tubular section is an elongate section.
- 19. (Amended) The method of claim 17, wherein the tubular section is a linear section.
- 20. (Amended) The method of claim 17, wherein the tubular section is substantially cylindrical.
- 21. (Amended) The method of claim 17, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 22. (Amended) The method of claim 17, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 23. (Amended) The method of claim 17, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 24. (Amended) The method of claim 1, wherein the aerosol flow is provided by entraining the aerosol in a flow of a carrier gas fed to the nozzle unit.
- 25. (Amended) The method of claim 1, wherein the aerosol flow is provided by applying a reduced pressure to the at least one outlet so as to entrain the aerosol in a flow of a carrier gas drawn through the nozzle unit.
- 26. (Amended) The method of claim 24, wherein the carrier gas is a gas reactive to the material solution.
- 27. (Amended) The method of claim 24, wherein the carrier gas is a gas non-reactive to the material solution.
- 28. (Amended) The method of claim 24 when appendant upon claim 4, wherein the flow of the carrier gas is provided such as to maintain the decreasing, temperature gradient.

- 29. (Amended) The method of claim 1, wherein the aerosol is delivered to the substrate such as to achieve a film growth rate of at least 0.2 um per minute, preferably at least 1 um per minute, more preferably at least 2 um per minute.
- 30. (Amended) The method of claim 1, wherein the flow rate through the at least one outlet is at least 5 ml per minute, preferably at least 50 ml per minute.
- 31. (Amended) The method of claim 1, wherein the nozzle unit is configured such that the directed aerosol flow from the at least one outlet is directed upwards, preferably substantially vertically upwards.
- 32. (Amended) The method of claim 1, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 33. (Amended) The method of claim 1, wherein the applied voltage is less than about 35 kV, preferably less than about 20 kV.
- 34. (Amended) The method of claim 1, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 35. (Amended) The method of claim 1, wherein the substrate is held stationary relative to the nozzle unit.
- 36. (Amended) The method of claim 1, further comprising the step of moving the nozzle unit relative to the substrate.
- 37. The method of claim 36, wherein the substrate is rotated, tilted and/or translated relative to the nozzle unit.
  - 38. (Amended) The method of claim 1, when performed at atmospheric pressure.
- 39. (Amended) The method of claim 1, when performed below atmospheric pressure.

- 40. (Amended) The method of claim 1, when performed above atmospheric pressure.
- 41. An apparatus for depositing material, preferably a film, on a substrate, comprising:

a substrate holder for holding a substrate;

a heater for heating the substrate;

an aerosol generator for generating an aerosol comprising droplets of a material

solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and

a high voltage supply for generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is in use attracted towards the substrate.

- 42. The apparatus of claim 41, where configured to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 43. (Amended) The apparatus of claim 41, wherein the at least one electrode extends upstream of the at least one outlet.
- 44. (Amended) The apparatus of claim 41, wherein the at least one electrode comprises an elongate element.
- 45. (Amended) The apparatus of claim 41, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.

- 46. (Amended) The apparatus of claim 41, wherein the distal end of the at least one electrode includes a single tip.
- 47. (Amended) The apparatus of claim 41, wherein the distal end of the at least one electrode includes a plurality of tips.
- 48. (Amended) The apparatus of claim 41, wherein the nozzle unit includes a tubular section upstream of each outlet.
  - 49. The apparatus of claim 48, wherein the tubular section is an elongate section.
- 50. (Amended) The apparatus of claim 48, wherein the tubular section is a linear section.
- 51. (Amended) The apparatus of claim 48, wherein the tubular section is substantially cylindrical.
- 52. (Amended) The apparatus of claim 48, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 53. (Amended) The apparatus of claim 48, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 54. (Amended) The apparatus of claim 48, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 55. (Amended) The apparatus of claim 41, further comprising a gas supply unit in communication with the aerosol generator for supplying a flow of a carrier gas for entraining the aerosol and delivering the same through the nozzle unit.
- 56. (Amended) The apparatus of claim 41, wherein the at least one outlet is directed upwards, preferably substantially vertically upwards.

- 57. (Amended) The apparatus of claim 41, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 58. (Amended) The apparatus of claim 41, wherein the nozzle unit and the substrate are held in fixed relation.
- 59. (Amended) The apparatus of claim 41, wherein the nozzle unit and the substrate are configured so as to be movable relative to one another.
- 60. The apparatus of claim 59, wherein the substrate is rotatable, tiltable and/or translatable relative to the nozzle unit.
- 63. (Amended) The apparatus of claim 41, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 64. A method of fabricating a powder, preferably an ultrafine powder, comprising the steps of:

providing a heated zone:

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to-form a powder.

65. An apparatus for fabricating a powder, preferably an ultrafine powder, comprising:

a heater for providing a heated zone;

an aerosol generator for generating an aerosol comprising droplets of a material solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and a high voltage supply for generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is in use attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to form a powder.

Please cancel claims 61 and 62 without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents.

# IN THE DRAWINGS

Please amend the drawings without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents as follows:

Page 4/7, please delete Fig. 7.

Page 5/7, please delete Fig. 8.

Page 5/7, please renumber Figure 9 as Figure 7.

Page 6/7, please renumber Figures 10(a) and (b) as Figures 8(a) and (b).

Page 7/7, please renumber Figures 11(a) and (b) as Figures 9(a) and (b).

### REMARKS

Only Kwang-Leong CHOY and Bo SU are named here as inventors because it is believed that Junfa MEI is not an inventor of the subject matter as now claimed.

This case has been amended to put it in better form for U.S. prosecution.

Please charge any additional fees incurred by reason of this paper to Deposit Account No. 50-0320.

An early examination on the merits is requested.

Attached hereto is a marked-up version of the changes made to the claims by the current Preliminary Amendment. The attached pages are captioned "Version with markings to show changes made".

Respectfully submitted,

FROMMER LAWRENCE & HAUG LLP

Bv: //

Thomas J. Kowalski

Reg. No. 32,147 (212) 588-0800

Enc. - Version with markings to show changes made

"Version with markings to show changes made"

Page 1, line 1:

This application is a Continuation-in-Part of PCT/GB00/00013, filed January 5, 2000, designating the U.S., published July 29, 2000 as WO 00/42234 and claiming priority from GB 9900955.7 filed January 15, 1999. All of the above-mentioned applications, as well as all documents cited herein and documents referenced or cited in documents cited herein, are hereby incorporated herein by reference

Page 9, paragraphs 9 and 10:

[Preferably, the apparatus further comprises a chamber for enclosing the substrate holder.

More preferably, the apparatus further comprises a further gas supply unit in communication with the chamber for separately delivering a further gas to the same.]

Page 11, last paragraph:

[Figure 7 schematically illustrates a film deposition apparatus in accordance with a third embodiment of the present invention.]

Page 12, first paragraph:

[Figure 8 illustrates a sectional view of the outlet end of a modified nozzle unit for the film deposition apparatus of Figure 7.]

Page 12, paragraph 2:

Figure [9] 7 illustrates an X-ray diffraction pattern of a BaZrO<sub>3</sub>; film as fabricated by Example 1;

Page 12, paragraph 3:

Figures [10] <u>8</u>(a) and (b) illustrate surface and cross-sectional SEM micrographs of a CdS film as fabricated by Example 2; and

Page 12, paragraph 4:

Figures [11]  $\underline{9}$ (a) and (b) illustrate surface and cross-sectional SEM micrographs of a porous SiO<sub>2</sub> film as fabricated by Example 3.

Page 18, please cancel lines 5 through 32.

[Figure 7 illustrates a film deposition apparatus in accordance with a third embodiment of the present invention which finds particular application in the deposition of films onto three-dimensional substrates of complex shape, such as tubular sections.

The film deposition apparatus comprises a heater 201 which includes an enclosed deposition chamber 202, in this embodiment an enclosed tue furnace, in which is provided a heated zone, and a substrate holder 203 for holding a substrate 205 in the heating chamber 202. The substrate holder 203 is rotatably disposed, in this embodiment about the longitudinal axis of the heating chamber 202, within the heater 201 such as to allow for the presentation of the entire surface of the substrate 205 to be coated to an aerosol flow. The heating chamber 202 includes an inlet port 206 through which a gas can be introduced thereinto, an outlet port 207 which acts as an exhaust, and a radial opening 208 through which a nozzle unit 211 extends for delivering an aerosol thereinto. In this embodiment the outlet port 207 is connected to a exhaust line 209 which includes a flow regulating valve 210 for regulating the flow rate from the heating chamber 202.

The film deposition apparatus further comprises a nozzle unit 211 which extends through the radial opening 208 in the deposition chamber 202 for delivering a directed aerosol flow to the substrate 205 to be coated. This nozzle unit 211 is of a similar construction to that employed in the film deposition apparatus of the above described first embodiment, with corresponding reference signs being used to designate like parts. The nozzle unit 211 comprises a tubular

section 215, in this embodiment an elongate cylindrical section with a inwardly-tapered outlet end, which includes an inlet port 217 at one end thereof through which an aerosol flow is introduced and an outlet port 218 at the other end thereof through which a directed aerosol flow is delivered to the substrate 205 to be coated, the internal geometry of the tubular section 215 being such as to confer directionality to the aerosol flow. The tubular section 215 is]

### Pages 19 through 21:

[preferably formed of a non-conductive, insulating material, such as a ceramic, glass of quartz, which can withstand the high temperatures developed by the heater 201. The nozzle unit 211 further comprises an electrode 221, in this embodiment an elongate element, such as a wire, having a single sharp-pointed tip, which extends co-axially through a length of the tubular section 215, in this embodiment with the tip thereof located upstream of the outlet port 218, that is, within the tubular section 215. The electrode 221 can be formed of any conductive material, but is preferably formed of aluminium, stainless steel, or tungsten. In a modified nozzle unit 211, as illustrated in Figure 5, the electrode 221 can be multi-tipped.

The film deposition apparatus further comprises an aerosol generator 225 for providing a flow of an aerosol to the inlet port 217 of the nozzle unit 211. The aerosol generator 225 comprises a chamber 227 which includes first and second inlet ports 229, 231 and an outlet port 233 connected by a flexible tubular section 234 to the inlet port 217 of the nozzle unit 211, and defines a reservoir 235 for containing a precursor solution 137 to be aerosolised and a head space 239 in which an aerosol collects when generated. The aerosol generator 225 further comprises a liquid level controller 241 connected by a line 242 to the first inlet port 229 of the chamber 227 for maintaining a constant volume of the precursor solution 237 in the reservoir 235. The aerosol generator 225 further comprises a piezoelectric transducer 243 which is driven by a

power supply 244 and is in communication with the reservoir 235 through a transfer medium 245, such as water, contained separately from the precursor solution 237 such that on operation of the piezoelectric transducer 243 the liquid precursor 237 is ultrasonically vibrated to generate an aerosol in the head space 239. The aerosol generator 225 further comprises a first gas supply unit 247 connected through a delivery line 249 to the second inlet port 231 of the chamber 227 for providing a flow of a carrier gas through the chamber 227 such as to entrain the aerosol in the head space 239 and transport the same to the substrate 205 through the nozzle unit 211. In this embodiment the first gas supply unit 247 includes a temperature conditioner for controlling the temperature of the delivered gas, the importance of which will become apparent hereinbelow. Further, in this embodiment the delivery line 249 includes a flow regulating valve 251 for controlling the gas flow rate and hence the flow rate of aerosol delivered to the substrate 205. Preferably, the carrier gas comprises at least one of air, Ar, H<sub>2</sub>S, N<sub>2</sub>, NH<sub>3</sub>, and O<sub>2</sub>. In an alternative embodiment, instead of or in addition to the first gas supply unit 247, pressure reducing means, such as a vacuum pump, could be provided for applying a reduced pressure at the outlet port 207 of the heating chamber 202 so as to draw the aerosol as a flow through the nozzle unit 211.

The film deposition apparatus further comprises a high voltage d.c. supply 253 connected between the electrode 221 and the substrate 205 such as to establish an electric field between the same, which electric field charges the aerosol droplets on passing the electrode 221 and causes the charged droplets to be attracted to the substrate 205 on exiting the outlet port 218 of the nozzle unit 211. In a preferred embodiment the voltage applied between the electrode 221 and the substrate 205 is from 10 to 30 kV. In this embodiment the film deposition apparatus further comprises a plurality of deflector plates 254 disposed with in the deposition chamber 202 about

the location of the substrate 205, which deflector plates 254 are connected to the high voltage d.c. supply 253 such as to have the same polarity as the electrode 221 and hence the charged aerosol droplets and act to deflect the aerosol droplets towards the substrate 205, thereby minimising the deposition of material on the internal walls of the deposition chamber 202.

The film deposition apparatus further comprises a second gas supply unit 255 connected by a delivery line 256 to the inlet port 206 of the deposition chamber 202 for providing a controlled environment in the deposition chamber 202. Preferably, the gas is an inert gas, such as argon or nitrogen. In this embodiment the delivery line 256 includes a flow regulating valve 257 for controlling the flow rate of gas delivered to the deposition chamber 202.

The film deposition apparatus further comprises a first motor unit 258 connected to the substrate holder 203 so as to provide for movement of the substrate holder 203 and hence the substrate 205, in this embodiment by rotation and axial movement, in the deposition chamber 202 relative to the outlet port 218 of the nozzle unit 211, a second motor unit 259 connected to the nozzle unit 211 so as to provide for movement of the nozzle unit 211, in this embodiment radial movement, in the deposition chamber 202 relative to the substrate holder 203 and hence the substrate 205, and a computer 261 for controlling the operation of the first and second motor units 258, 259. The distance between the outlet port 218 of the nozzle unit 211 and the surface of the substrate 205 being coated is preferably maintained at less than 100mm, more preferably less than 50 mm, and still more preferably not more than 20 mm.

In use, the aerosol generator 225 is operated to provide a gas flow entraining aerosol droplets through the nozzle unit 211, which flow through the nozzle unit 211 provides a directed aerosol flow from the outlet prot 218 of the tubular section 215 and results in charging of the aerosol droplets on passing the electrode 221. On exiting the outlet port 218 of the nozzle unit

211 the charged aerosol droplets are attracted to the substrate 205, with the flow rate of the aerosol, and the temperature and temperature gradient at the surface of the substrate 205 being optimised to achieve the desired film properties, typically one of a porous or dense solid film. In this embodiment a temperature gradient is maintained at the surface of the substrate 205 by controlling both the temperature and the flow rate of the carrier gas supplied by the first gas supply unit 247. In preferred embodiments the thermal environment and the velocity of the directed aerosol flow can be configured such that the aerosol droplets are vaporised/decomposed close to the surface of the substrate 205 or impact the surface of the substrate 205 prior to vaporisation/decomposition. With the continued relative movement of the substrate 205 and the nozzle unit 211, this process is continued until a film of the required thickness has been achieved over the surface of the substrate 205.

In one modification to the above-described third embodiment, as illustrated in Figure 8, the nozzle unit 211 can include a cooling jacket 261 located about the tubular section 215 for cooling the internal volume of the tubular section 215 through which the aerosol is delivered. The cooling jacket 261 includes a cavity 263 through which a cooling medium, typically a liquid, such as oil or water, is continuously circulated. The nozzle unit 211 can also incude a secondary electrode 265, in this embodiment an annular element located about the longitudinal axis of the primary electrode 221,]

Page 22:

[disposed downstream of the primary electrode 221 which acts to focus and accelerate the aerosol droplets towards the substrate 205.

In another modification to the above-described third embodiment, the apparatus can be set up to fabricate powders, preferably ultrafine powders. In this modification, the substrate

holder 203 and the deflector plates 254 are removed and replaced by a plate which is disposed substantially opposite to the outlet port 218 of the nozzle unit 211 and connected to the high d.c. supply 253 such that an electric field is developed between the plate and the electrode 221. In use, with the thermal environment in the heated zone configured appropriately, the aerosol droplets exiting the nozzle unit 211 react homogeneously in the gas phase to provide a powder which collects in the heating chamber 202. By controlling the size of the aerosol droplets ultrafine powders can be fabricated.]

Page 22 to 23, last paragraph:

A non-aqueous precursor solution for the deposition of a BaZrO<sub>3</sub> film was first prepared as follows. Barium metal (as supplied by Aldrich) was completely dissolved in a volume of 2-methoxyethanol (as supplied by Aldrich) by stirring at room temperature to form a barium alkoxide solution. A stoichiometric amount of zirconium n-propoxide, a 70 wt% solution in n-propanol (as supplied by Aldrich), was then added to the barium methoxyoxide solution and refluxed at 124 °C, the boiling point of 2-methoxyethanol, for five hours. Then, a volume of 2-methoxyethanol was added to the refluxed solution to provide a 0.05 M precursor solution. Using the apparatus of the first-described embodiment and the so-prepared solution, a BaZrO<sub>3</sub> film was deposited on a silver substrate 5, with a substrate temperature of 600 °C, a substrate 5 to nozzle unit 11 distance of 30 mm, an electric field voltage of 10 kV, the piezoelectric transducer 43 of the aerosol generator 25 being operated at a frequency of 1.7 MHz and power of 50 W, and nitrogen being supplied at 30 ml per minute as the carrier gas. Nitrogen was used as the carrier gas to minimise the reaction between the barium and carbon dioxide in the air. The resulting film. formed in a single run without the need for any post-deposition heat treatment, was a

crystalline BaZrO<sub>3</sub> film as characterized by the X-ray diffraction pattern illustrated in Figure (9) 7.

Page 23, the first full paragraph:

A 0.01 M aqueous precursor solution for the deposition of a CdS film was first prepared using cadmium chloride and thiourea. Using the apparatus of the second-described embodiment and the so-prepared solution, a CdS film was deposited on a glass substrate 105, with a substrate temperature of 450 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 50 W, a deposition time of five minutes, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a dense, crystalline CdS film having a thickness of about 1 um, with a columnar structure and a smooth and uniform surface. SEM micrographs of the resulting film are illustrated in Figures [10] 8(a) and (b). --.

Page 23, the second full paragraph:

A colloidal silica solution (Ludox<sup>TM</sup>, as supplied by DuPont) was diluted with distilled water to prepare an aqueous precursor solution having a concentration of 0.1 g/ml for the deposition of a SiO<sub>2</sub> film. Using the apparatus of the second-described embodiment and the soprepared solution, a SiO<sub>2</sub> film was deposited on a glass substrate 105, with a substrate temperature of 200 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 20 W, a deposition time of one minute, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without

the need for any post-deposition heat treatment, was a porous  $SiO_2$  film with a reticular structure. SEM micrographs of the resulting film are illustrated in Figures [11] 9(a) and (b). --.

Page 24, first paragraph:

Finally, it will be understood that the present invention has been described in its preferred embodiments and can be modified in many different ways within the scope of the invention as defined by the appended claims. For example, in coating substrates 5, 105[, 205] of large area or complex geometric shape, the nozzle units 11, 111[, 211] could be modified to include a plurality of outlet ports 18, 118[, 218] or the film deposition apparatus could be modified to include a plurality of nozzle units 11, 111[, 211].

Page 24, after the first paragraph:

The invention will now be further described by the following numbered paragraphs.

1. A method of depositing material, preferably a film, on a substrate, comprising the steps of:

providing a substrate;

heating the substrate;

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

- generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is attracted towards the substrate.
- 2. The method of paragraph 1, wherein the substrate is heated to a temperature of less than about 1050 °C, preferably less than about 800 °C.
- 3. The method of paragraph 1 or 2, wherein the substrate is heated during deposition,
- 4. The method of paragraph 3, wherein the thermal environment is such as to

  maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 5. The method of any of paragraphs 1 to 4, wherein the material solution is an aqueous solution.
- 6. The method of any of paragraphs 1 to 4, wherein the material solution is a non-aqueous solution.
- 7. The method of any of paragraphs 1 to 6, wherein the aerosol droplets are at least partially charged prior to exiting the at least one outlet.
- 8. The method of paragraph 7, wherein the aerosol droplets are charged prior to exiting the at least one outlet.
- 9. The method of any of paragraphs 1 to 7, wherein the aerosol droplets are at least partially charged after exiting the at least one outlet.
- 10. The method of any of paragraphs 1 to 9, wherein the aerosol droplets are charged by the at least one electrode.
- 11. The method of any of paragraphs 1 to 10, wherein the at least one electrode is disposed at least partially in each aerosol flow.

- 12. The method of any of paragraphs 1 to 11, wherein the at least one electrode extends upstream of the at least one outlet.
- 13. The method of any of paragraphs 1 to 12, wherein the at least one electrode comprises an elongate element.
- 14. The method of any of paragraphs 1 to 13, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 15. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a single tip.
- 16. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a plurality of tips.
- 17. The method of any of paragraphs 1 to 16. wherein the nozzle unit includes a tubular section upstream of each outlet.
- 18. The method of paragraph 17. wherein the tubular section is an elongate section.
- 19. The method of paragraph 17 or 18, wherein the tubular section is a linear section.
- 20. The method of any of paragraphs 17 to 19, wherein the tubular section is substantially cylindrical.
- 21. The method of any of paragraphs 17 to 20, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 22. The method of any of paragraphs 17 to 21, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 23. The method of any of paragraphs 1 to 22, wherein at least the inner surface of the tubular section is composed of an insulating material.

- 24. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by entraining the aerosol in a flow of a carrier gas fed to the nozzle unit.
- 25. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by applying a reduced pressure to the at least one outlet so as to entrain the aerosol in a flow of a carrier gas drawn through the nozzle unit.
- 26. The method of paragraph 24 or 25, wherein the carrier gas is a gas reactive to the material solution.
- 27. The method of paragraph 24 or 25, wherein the carrier gas is a gas non-reactive to the material solution.
- 28. The method of any of paragraphs 24 to 27 when appendant upon paragraph 4,

  wherein the flow of the carrier gas is provided such as to maintain the decreasing,
  temperature gradient.
- 29. The method of any of paragraphs 1 to 28, wherein the aerosol is delivered to the substrate such as to achieve a film growth rate of at least 0.2 um per minute, preferably at least 1 um per minute, more preferably at least 2 um per minute.
- 30. The method of any of paragraphs 1 to 29, wherein the flow rate through the at least one outlet is at least 5 ml per minute, preferably at least 50 ml per minute.
- 31. The method of any of paragraphs 1 to 30, wherein the nozzle unit is configured such that the directed aerosol flow from the at least one outlet is directed upwards, preferably substantially vertically upwards.
- 32. The method of any of paragraphs 1 to 31, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.

- 33. The method of any of paragraphs 1 to 32, wherein the applied voltage is less than about 35 kV, preferably less than about 20 kV.
- 34. The method of any of paragraphs 1 to 33, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 35. The method of any of paragraphs 1 to 34, wherein the substrate is held stationary relative to the nozzle unit.
- 36. The method of any of paragraphs 1 to 34, further comprising the step of moving the nozzle unit relative to the substrate.
- The method of paragraph 36, wherein the substrate is rotated, tilted and/or translated relative to the nozzle unit.
- 38. The method of any of paragraphs 1 to 37, when performed at atmospheric pressure.
- 39. The method of any of paragraphs 1 to 37, when performed below atmospheric pressure.
- 40. The method of any of paragraphs 1 to 37, when performed above atmospheric pressure.
- 41. An apparatus for depositing material, preferably a film, on a substrate, comprising:
  - a substrate holder for holding a substrate;
  - a heater for heating the substrate;
  - an aerosol generator for generating an aerosol comprising droplets of a material solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the
aerosol to the substrate, the nozzle unit including at least one outlet through which
a directed flow of the aerosol is in use delivered and at least one electrode; and
a high voltage supply for generating an electric field between the substrate and the
at least one electrode such that the directed aerosol flow is in use attracted
towards the substrate.

- 42. The apparatus of paragraph 41, where configured to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 43. The apparatus of paragraph 41 or 42, wherein the at least one electrode extends upstream of the at least one outlet.
- 44. The apparatus of any of paragraphs 41 to 43, wherein the at least one electrode comprises an elongate element.
- 45. The apparatus of any of paragraphs 41 to 44, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 46. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a single tip.
- 47. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a plurality of tips.
- 48. The apparatus of any of paragraphs 41 to 47, wherein the nozzle unit includes a tubular section upstream of each outlet.

- 49. The apparatus of paragraph 48, wherein the tubular section is an elongate section.
- 50. The apparatus of paragraph 48 or 49, wherein the tubular section is a linear section.
- 51. The apparatus of any of paragraphs 48 to 50, wherein the tubular section is substantially cylindrical.
- 52. The apparatus of any of paragraphs 48 to 51, wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 53. The apparatus of any of paragraphs 48 to 52, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 54. The apparatus of any of paragraphs 48 to 53, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 55. The apparatus of any of paragraphs 41 to 54, further comprising a gas supply unit in communication with the aerosol generator for supplying a flow of a carrier gas for entraining the aerosol and delivering the same through the nozzle unit.
- 56. The apparatus of any of paragraphs 41 to 55, wherein the at least one outlet is directed upwards, preferably substantially vertically upwards.
- 57. The apparatus of any of paragraphs 41 to 56, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 58. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are held in fixed relation.
- 59. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are configured so as to be movable relative to one another.

- 60. The apparatus of paragraph 59, wherein the substrate is rotatable, tiltable and/or translatable relative to the nozzle unit.
- 61. The apparatus of any of paragraphs 41 to 60, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 62. A method of fabricating a powder, preferably an ultrafine powder, comprising the steps of:

providing a heated zone:

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed

flow of the aerosol from the at least one outlet; and

generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to-form a powder.

An apparatus for fabricating a powder, preferably an ultrafine powder, comprising:

a heater for providing a heated zone;

an aerosol generator for generating an aerosol comprising droplets of a material solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the

aerosol to the heated zone, the nozzle unit including at least one outlet through

which a directed flow of the aerosol is in use delivered and at least one electrode;

and

a high voltage supply for generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is in use attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to form a powder.

## IN THE CLAIMS:

1. A method of depositing material, preferably a film, on a substrate, comprising the steps of:

providing a substrate;

heating the substrate;

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is attracted towards the substrate.

- 2. The method of claim 1, wherein the substrate is heated to a temperature of less than about 1050 °C, preferably less than about 800 °C.
- 3. (Amended) The method of claim 1 [or 2], wherein the substrate is heated during deposition,
- 4. The method of claim 3, wherein the thermal environment is such as to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 5. (Amended) The method of [any of] claim[s] 1 [to 4], wherein the material solution is an aqueous solution.
- 6. (Amended) The method of [any of] claim[s] 1 [to 4], wherein the material solution is a non-aqueous solution.
- 7. (Amended) The method of [any of] claim[s] 1 [to 6], wherein the aerosol droplets are at least partially charged prior to exiting the at least one outlet.
- 8. The method of claim 7, wherein the aerosol droplets are charged prior to exiting the at least one outlet.
- 9. (Amended) The method of [any of] claim[s] 1 [to 7], wherein the aerosol droplets are at least partially charged after exiting the at least one outlet.
- 10. (Amended) The method [of any] of claim[s] 1 [to 9], wherein the aerosol droplets are charged by the at least one electrode.
- 11. (Amended) The method of [any of] claim[s] 1 [to 10], wherein the at least one electrode is disposed at least partially in each aerosol flow.

- 12. (Amended) The method of [any of] claim[s] 1 [to 11], wherein the at least one electrode extends upstream of the at least one outlet.
- 13. (Amended) The method of [any of] claim[s] 1 [to 12], wherein the at least one electrode comprises an elongate element.
- 14. (Amended) The method of [any of] claim[s] 1 [to 13], wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 15. (Amended) The method of [any of] claim[s] 1 [to 14], wherein the distal end of the at least one electrode includes a single tip.
- 16. (Amended) The method of [any of] claim[s] 1, [to 14], wherein the distal end of the at least one electrode includes a plurality of tips.
- 17. (Amended) The method of [any of] claim[s] 1 [to 16.], wherein the nozzle unit includes a tubular section upstream of each outlet.
- 18. (Amended) The method of claim 17[.], wherein the tubular section is an elongate section.
- 19. (Amended) The method of claim 17 [or 18], wherein the tubular section is a linear section.
- 20. (Amended) The method of [any of] claim[s] 17 [to 19], wherein the[.] tubular section is substantially cylindrical.
- 21. (Amended) The method of [any of] claim[s] 17 [to 20], wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 22. (Amended) The method of [any of] claim[s] 17 [to 21], wherein the at least one electrode extends substantially along the central axis of the associated tubular section.

- 23. (Amended) The method of [any of] claim[s] [1 to 22]17, wherein at least the inner surface of the tubular section is composed of an insulating material.
- 24. (Amended) The method of [any of] claim[s] 1 [to 23], wherein the aerosol flow is provided by entraining the aerosol in a flow of a carrier gas fed to the nozzle unit.
- 25. (Amended) The method of [any of] claim[s] 1 [to 23], wherein the aerosol flow is provided by applying a reduced pressure to the at least one outlet so as to entrain the aerosol in a flow of a carrier gas drawn through the nozzle unit.
- 26. (Amended) The method of claim 24 [or 25], wherein the carrier gas is a gas reactive to the material solution.
- 27. (Amended) The method of claim 24 [or 25], wherein the carrier gas is a gas non-reactive to the material solution.
- 28. (Amended) The method of [any of] claim[s] 24 [to 27] when appendant upon claim 4, wherein the flow of the carrier gas is provided such as to maintain the decreasing, temperature gradient.
- 29. (Amended) The method of [any of] claim[s] 1 [to 28], wherein the aerosol is delivered to the substrate such as to achieve a film growth rate of at least 0.2 um per minute, preferably at least 1 um per minute, more preferably at least 2 um per minute.
- 30. (Amended) The method of [any of] claim[s] 1 [to 29], wherein the flow rate through the at least one outlet is at least 5 ml per minute, preferably at least 50 ml per minute.
- 31. (Amended) The method of [any of] claim[s] 1 [to 30], wherein the nozzle unit is configured such that the directed aerosol flow from the at least one outlet is directed upwards, preferably substantially vertically upwards.

- 32. (Amended) The method of [any of] claim[s] 1 [to 31], wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 33. (Amended) The method of [any of] claim[s] 1 [to 32], wherein the applied voltage is less than about 35 kV, preferably less than about 20 kV.
- 34. (Amended) The method of [any of] claim[s] 1 [to 33], wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 35. (Amended) The method of [any of] claim[s] 1 [to 34], wherein the substrate is held stationary relative to the nozzle unit.
- 36. (Amended) The method of [any of] claim[s] 1 [to 34], further comprising the step of moving the nozzle unit relative to the substrate.
- 37. The method of claim 36, wherein the substrate is rotated, tilted and/or translated relative to the nozzle unit.
- 38. (Amended) The method of [any of] claim[s] 1 [to 37], when performed at atmospheric pressure.
- 39. (Amended) The method of [any of] claim[s] 1 [to 37], when performed below atmospheric pressure.
- 40. (Amended) The method of [any of] claim[s] 1 [to 37], when performed above atmospheric pressure.
- 41. An apparatus for depositing material, preferably a film, on a substrate, comprising:
  - a substrate holder for holding a substrate;
  - a heater for heating the substrate;

an aerosol generator for generating an aerosol comprising droplets of a material solution;

a charge applicator for applying a positive or negative charge to the aerosol droplets;

a nozzle unit in communication with the aerosol generator for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and

a high voltage supply for generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is in use attracted towards the substrate.

- 42. The apparatus of claim 41, where configured to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
- 43. (Amended) The apparatus of claim 41 [or 42], wherein the at least one electrode extends upstream of the at least one outlet.
- 44. (Amended) The apparatus of [any of] claim[s] 41 [to 43], wherein the at least one electrode comprises an elongate element.
- 45. (Amended) The apparatus of [any of] claim[s] 41 [to 44], wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
- 46. (Amended) The apparatus of [any of] claim[s] 41 [to 45], wherein the distal end of the at least one electrode includes a single tip.
- 47. (Amended) The apparatus of [any of] claim[s] 41 [to 45], wherein the distal end of the at least one electrode includes a plurality of tips.
- 48. (Amended) The apparatus of [any of] claim[s] 41 [to 47], wherein the nozzle unit includes a tubular section upstream of each outlet.

- 49. The apparatus of claim 48, wherein the tubular section is an elongate section.
- 50. (Amended) The apparatus of claim 48 [or 49], wherein the tubular section is a linear section.
- 51. (Amended) The apparatus of [any of] claim[s] 48 [to 50], wherein the tubular section is substantially cylindrical.
- 52. (Amended) The apparatus of [any of] claim[s] 48 [to 51], wherein the at least one electrode extends substantially entirely through the associated tubular section.
- 53. (Amended) The apparatus of [any of] claim[s] 48 [to 52], wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
- 54. (Amended) The apparatus of [any of] claim[s] 48 [to 53], wherein at least the inner surface of the tubular section is composed of an insulating material.
- 55. (Amended) The apparatus of [any of] claim[s] 41 [to 54], further comprising a gas supply unit in communication with the aerosol generator for supplying a flow of a carrier gas for entraining the aerosol and delivering the same through the nozzle unit.
- 56. (Amended) The apparatus of [any of] claim[s] 41 [to 55], wherein the at least one outlet is directed upwards, preferably substantially vertically upwards.
- 57. (Amended) The apparatus of [any of] claim[s] 41 [to 56], wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
- 58. (Amended) The apparatus of [any of] claim[s] 41 [to 57], wherein the nozzle unit and the substrate are held in fixed relation.
- 59. (Amended) The apparatus of [any of] claim[s] 41 [to 57], wherein the nozzle unit and the substrate are configured so as to be movable relative to one another.

- 60. The apparatus of claim 59, wherein the substrate is rotatable, tiltable and/or translatable relative to the nozzle unit.
- (Amended) [61. The apparatus of any of claims 41 to 60, further comprising a chamber for enclosing the substrate holder.)
- (Amended) [62. The apparatus of claim 61, further comprising a further gas supply unit in communication with the chamber for separately delivering a further gas to the same.]
- 63. (Amended) The apparatus of [any of] claim[s] 41 [to 62], wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
- 64. A method of fabricating a powder, preferably an ultrafine powder, comprising the steps of:

providing a heated zone:

generating an aerosol comprising droplets of a material solution;

providing a nozzle unit for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;

charging the aerosol droplets with a positive or negative charge;

providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to-form a powder.

65. An apparatus for fabricating a powder, preferably an ultrafine powder, comprising:

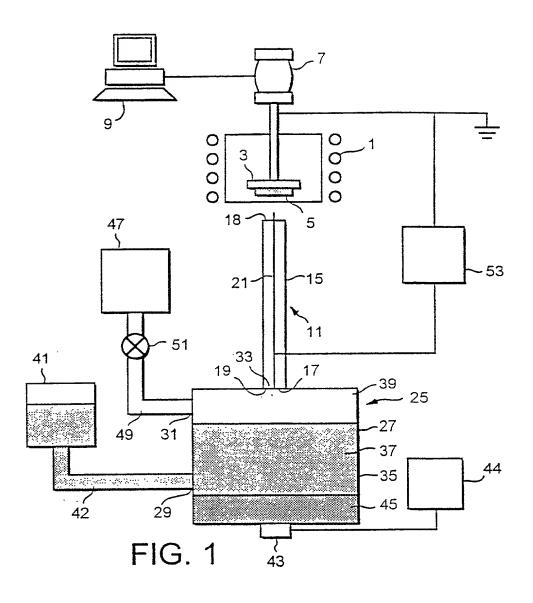
a heater for providing a heated zone;

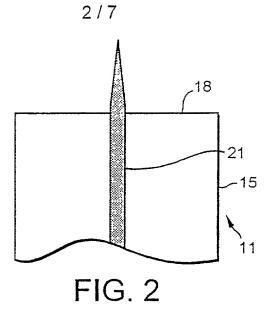
an aerosol generator for generating an aerosol comprising droplets of a material solution;

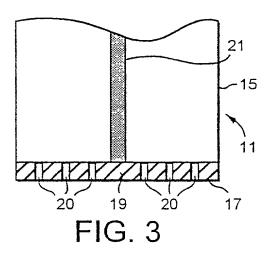
a charge applicator for applying a positive or negative charge to the aerosol droplets;

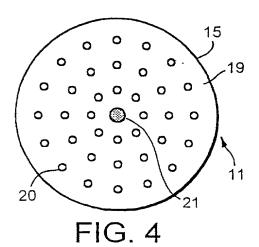
a nozzle unit in communication with the aerosol generator for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and

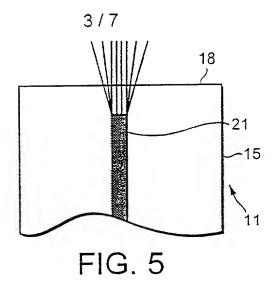
a high voltage supply for generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is in use attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to form a powder.

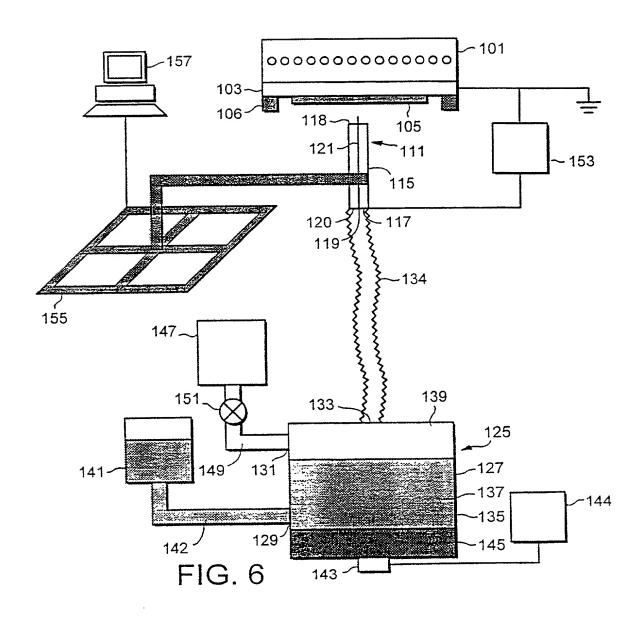




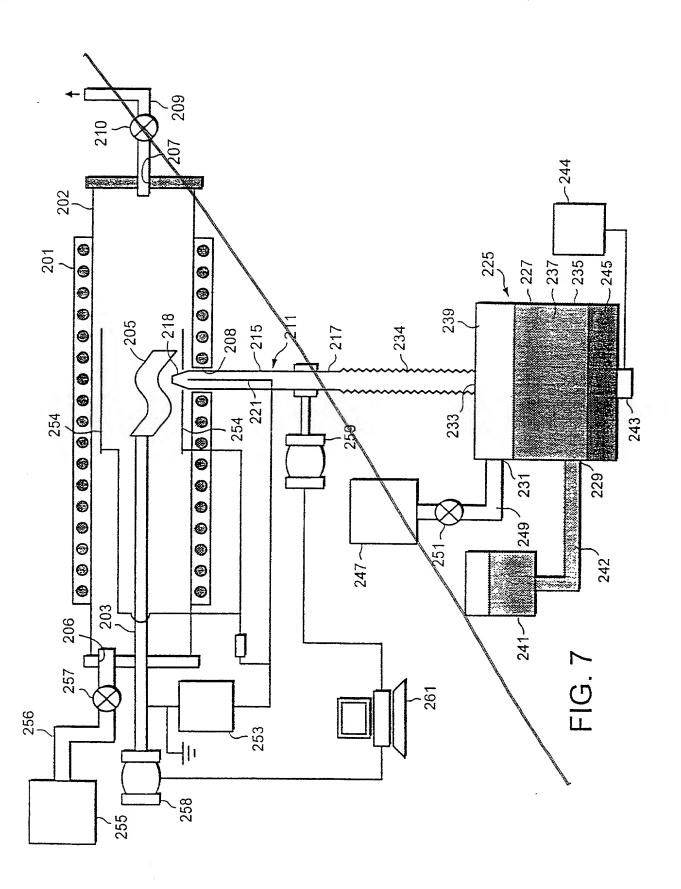




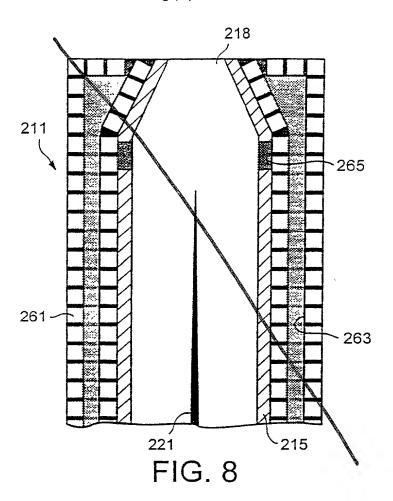


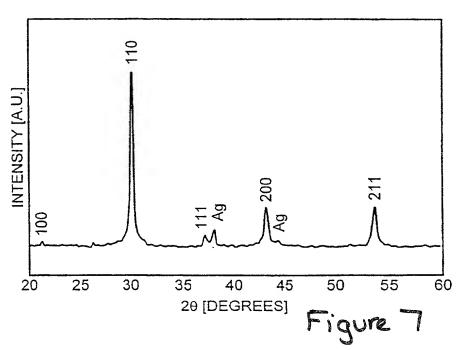


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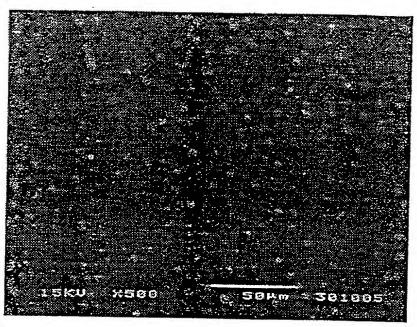


FIG. 10(a)

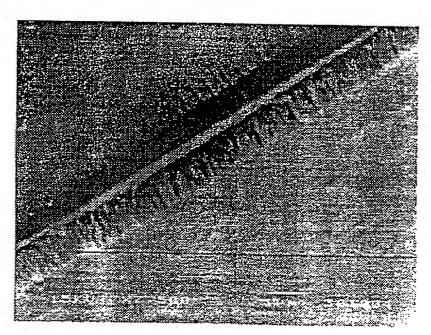


FIG. 10(b)

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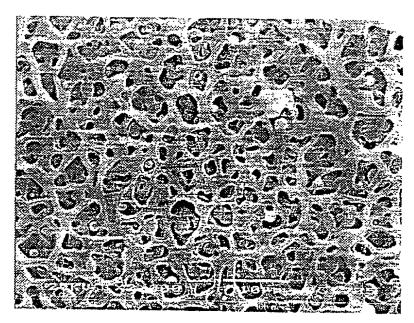


FIG. 1/(a)

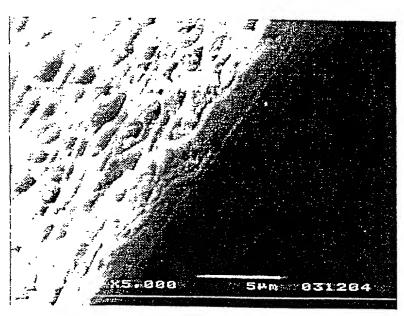


FIG. 1/1(b)

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